

## **High-precision uranium isotopic analysis for environmental forensics using MC-ICPMS: Demonstration studies at the Hanford Site, Washington**

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The contrasts in isotopic composition between natural and anthropogenic uranium and the wide variation in the composition of different processed uranium sources, promotes the measurement of uranium isotopic composition as a fingerprint and tracer of uranium contamination in the environment. Previous studies mainly have focused on the use of only one of the isotope ratios of U, e.g.  $^{234}\text{U}/^{238}\text{U}$ ,  $^{238}\text{U}/^{235}\text{U}$  or  $^{236}\text{U}/^{238}\text{U}$ . We measure all three of these ratios in environmental samples in order to better distinguish, characterize, identify and apportion U sources. For our U isotopic measurements, we employ an IsoProbe (GV Instr. Inc.) multiple-collector ICP source magnetic sector mass spectrometer. U isotopic compositions are measured simultaneously using a combination of Faraday cups (for  $^{235}\text{U}$  and  $^{238}\text{U}$ ) and a Daly photomultiplier ion counting system (for  $^{234}\text{U}$  and  $^{236}\text{U}$  in two separate analyses). U is separated from samples (e.g. vadose zone pore water, groundwater, rock/soil samples) prior to introduction to the MC-ICPMS via a desolvation system. At 7E-11 amps of  $^{238}\text{U}$  ions, a single analysis of a 20ppb U solution uses ~10ng of sample U. For correction of instrumental mass fractionation, we use bracketing analyses of a natural secular equilibrium U standard. This allows us to avoid the use of a double  $^{233}\text{U}$ - $^{236}\text{U}$  spike for mass fractionation correction that would compromise our ability to measure  $^{236}\text{U}/^{238}\text{U}$ . We also use the standard analyses for Daly/Faraday gain and for peak-tail correction of the  $^{236}\text{U}$  analyses. Typical precision for  $^{238}\text{U}/^{235}\text{U}$  is  $\leq 0.05\%$  2s, while for  $^{234}\text{U}/^{238}\text{U}$  it is  $\leq 0.15\%$  2s. Precision for  $^{236}\text{U}/^{238}\text{U}$  is  $\leq 0.15\%$  2s down to the  $10^{-7}$  range where precision degrades by a factor of ten. The limit for  $^{236}\text{U}/^{238}\text{U}$  measurement is about  $2 \times 10^{-8}$ , only ~five times higher than accelerator MS. For 1ppb U, this represents  $5 \times 10^7$  atoms  $^{236}\text{U}$  per liter water.

We will present three ongoing studies at the Hanford site as a demonstration of our techniques: (1) investigation of groundwater and vadose zone contamination in the B-BX-BY WMA (Christensen et al. (2004) Env. Sci. Tech., in press) (2) signatures of vadose zone contamination in waste cribs near U-Plant and (3) investigation of U contamination of the Columbia River.

New approach and technique for tracing uranium in the environment.